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RAdial DIstribution Least-Squares Program (RADILS)

Users Manual

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Laboratory for the Structure of Matter

October 17, 1979

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A rapidly converging radial distribution least squares program (RADILS) is available for the analysis of x-ray, neutron and electron diffraction data from liquid and amorphous materials. The program automatically generates a radial distribution function, G(r), essentially free from termination effects and determines the empirical background scale and shape, absolute scale factor, bulk density and coordination numbers, disorder parameters and interatomic distances for the first few highly ordered distances in the sample.	

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RADIAL DISTRIBUTION LEAST-SQUARES PROGRAM (RADILS)

USERS MANUAL

I. PRINCIPLES BEHIND THE APPROACH

Structural information is derived from the distribution of distances G(r) (EQ 1) in the sample, which is the Fourier sine transform of the interatomic interference intensity si(s). The momentum transfer s equals $4\pi\sin\theta/\lambda$ where λ is the wavelength and 2θ is the angle between the incident and diffracted beams. si(s) is not directly observable experimentally. The total diffracted intensity $I_t(s)$ (EQ 2) is composed of the interference intensity, I(s), coherent and incoherent atomic intensities, $I_a(s)$, plus multiple, $I_m(s)$, and extraneous, $I_e(s)$, scattering terms. After subtracting the non-interference terms, which we collectively refer to as the background scattering, B(s) (EQS 2 and 3), and divide by a sharpening function, generally Σf^2, we generate the interference intensity i(s) (EQ 4) corresponding approximately to the intensity from vibrating point atoms.

To generate an accurate G(r) from a truncated set of total intensity data, $I_t(s)$, a data reduction procedure must:

1. Isolate si(s) from $I_t(s)$ by determing the experimental background intensity B(s).
2. Minimize errors due to termination and scaling of the intensity data.

The radial distribution least-squares program (RADILS) described here is characterized by a unique treatment of termination effects, wherein the contributions from the first few ordered distances in the sample are subtracted from si(s) to form si'(s) (EQ 5), before the Fourier transform which generates G'(r) (EQ 6) is performed. G'(r) is expressed explicity (EQ 7) as a function of the total intensity, scattering factors and refinable parameters for the scale and shape of B(s), the absolute intensity scale factor, the bulk density, ρ_o, and the coordination numbers, disorder parameters and interatomic distances for the first few ordered distances. Equation 10 is minimized subject to the following constraints:

1. The inner region of $G'(r) + 4\pi\rho_o$ should be zero where distances are known not to exist. By minimizing termination effects

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which affect this region as well as the rest of G'(r), this region can be utilized as a sensitive measure of background error.

2. The outer region of G'(r) should show a uniform distribution of distances.

3. The scaling of the intensity data should be consistent with the bulk density and/or known coordination numbers.

4. B(s) should be a smoothly changing function whose shape is compatible with theory.

This approach has several advantages:

1. Termination error is minimized because si'(s) is essentially terminated at the experimental data limit s_{max}. This is the case because the primary contributors beyond s_{max} are due to the first few ordered distances because of their low disorder parameters.

2. The structural parameters for the short distances can be accurately determined by minimizing the spurious detail in the region of G'(r) where these distances contribute. The range over which $G'(r) + 4\pi r\rho_o$ is required to be zero is extended to include the region involving the distances which are subtracted.

3. Errors in the structural parameters of the ordered distances arising from the variability of the scattering factors with s, for x-rays and electrons, are minimized because the short distance contributions, with variable scattering factors, are subtracted before transformation.

G(r) is generated by reintroducing a gaussian description of the short distances without the associated termination effects.

Further details can be found in the following references:

1. J.H. Konnert and J. Karle, Acta Cryst. (1973) A29, 702.
2. P. D'Antonio, P. Moore, J.H. Konnert and J. Karle, Transactions of the American Crystallographic Association (1977), 13, 43.
3. K.N. Raymond, Lecture Notes, Least-Squares Tutorial, Spring Meeting, American Crystallographic Association, Berkeley, CA, 1974.

$$G(r) = 4\pi r(\rho(r) - \rho_o) = \frac{2}{\pi}\int_0^\infty si(s)\sin sr\,ds \qquad (1)$$

$$I_t(s) = I(s) + I_a(s) + I_m(s) + I_e(s) \qquad (2)$$

$$I_t(s) = I(s) + B(s) \qquad (3)$$

$$i(s) = (I_t(s) - B(s))/\Sigma f^2 \qquad (4)$$

$$si(s) = si(s) - SK\sum_{sd} N_{AB}C_{AB}\sin(sr_{AB})/r_{AB}\exp(-l_{AS}^2 s^2/2) \qquad (5)$$

sd refers to the first few ordered short distances.

For electron diffraction $C_{AB} = f_A(s)f_B(s)\cos(\eta_A(s) - \eta_B(s)/\Sigma f^2$

for x-ray and neutron diffraction $\eta = 0$.

$$G'(r) = 4\pi r(\rho'(r) - \rho_o) = \frac{2}{\pi}\int_0^{smax} si'(s)\sin sr\,ds \qquad (6)$$

$$G'(r) = SKr\rho_o + \frac{2}{\pi}\left[\sum_s s(I_t(s) - B(s))/\Sigma f^2 - SK\sum_{sd} N_{AB}C_{AB}\sin sr_{AB}\exp[-l_{AB}^2 s^2/2)/r_{AB}]\exp(-as^2)\sin sr\Delta s\right. \qquad (7)$$

B(s) can be expressed two ways in the program:

1. OVERLAPPING EXPONENTIAL BACKGROUND - single and multiple range data

$$B_k(s) = SK5_k[Ramp(s) + \sum_{n=1}^{NCV(k)} (W_{km}(s)\exp(A_{km} + B_{km}s^{C_{km}} + D_{km}s)] \qquad (8)$$

NCV(k) are the number of background segments in the k^{th} background for the k^{th} overlapping set of data. For single range data k = 1.

2. GAUSSIAN SUMMATION BACKGROUND - single range data only

$$B(s) = SK5\left(\sum_{i=1}^{NG} A_i \exp(-B_i s^2) + CO + Ramp(s)\right) \tag{9}$$

NG equals the number of gaussians

$Ramp(s) = FA.s$

$$\text{Minimize} \left\{ \sum_{\substack{\text{inner} \\ \text{region}}} (G'(r) + 4\pi r\rho_o)^2 + w\sum (G'(r))^2 \right\} \tag{10}$$

II. SCOPE

The Radial Distribution Least-Squares Program (RADILS) can be used to analyze amorphous or liquid x-ray, neutron or electron diffraction data, by providing the appropriate scattering factors. X-ray and neutron diffraction scattering coefficients are calculated in RADILS while those for electron diffraction must be supplied from a separate program. A choice among several background treatments and solutions of the normal equations is provided. The structural parameters can be constrained with linear constraint relations if necessary.

Polycrystalline data can be broadened to simulate amorphous scattering for topology testing by RDF comparison.

RADILS has been programmed to process up to four sets of overlapping data covering different data ranges, but this version has not been extensively tested.

This situation is common in the sector-microdensitometer method of electron diffraction and for x-ray data where different slit systems are used to optimize counting statistics.

The approach introduces additional parameters into the refinement, i.e. a scale factor for each range of data and restraints which restrain the interference intensities, in the overlap regions, to agree within specified limits. Alternatively, the end points of the reduced intensities, in the overlap region, can be constrained to be equal. This approach allows each data range its own background curve.

III. INPUT

A. Control section (required)

CONTROL CARD 1 (20A4)

COLS.	PARAMETER	DESCRIPTION
1-68	MTITL(1) - MTITL(17)	Descriptive title card
69-72	MTITL(18) = DF43 (Literal)	Scattering factors are read from file 43
	MTITL(18) = blank	Scattering factors read from cards
73-76	MTITL(19)= DF42 (Literal)	Experimental data are read from file 42.
	MTITL(19) = blank	Experimental data are read from cards
77-80	MTITL(20)= XRTI (Literal)	X-ray or neutron data are read in XRTI and processed
	MTITL(20)= EDIL (Literal)	Electron diffraction data are read in EDIL and processed

CONTROL CARD 2 (40I2)

1-2	IDX(1)<0	No DSIS plot
3-4	IDX(2)<0	No CSIS plot
5-6	IDX(3)<0	Intensities will be broadened
7-8	IDX(4)<0	Print all matrices during refinement
9-10	IDX(5)<0	G'(r) is calculated only to 6 A and rG(r) is not plotted at all. Used in the initial stages of refinement.
11-12	IDX(6)<0	Eliminates echo print of input intensities
13-14	IDX(7)<0	No back transform of $\Delta G'(r)$ is calculated.
15-16	IDX(8)<0	No print of s, TI, SIS, σ(SIS), r, rG(r) and σ(rG(r)) after last cycle.

COLS.	PARAMETER	DESCRIPTION
17-18	IDX(9)<0	Used for multiple data ranges. Only the beginning of each overlapping reduced intensity range is constrained to be the same.
	IDX(9)>0	Both beginning and end of each intensity overlap region are constrained to be the same. Not applicable for single range data NCAS = 1.
19-20	IDX(10)<0	No constraints on the reduced intensity overlap regions at all. Not applicable for single range data.
21-22	IDX(11)<0	SK3(NCAS) - the scale of the last overlapping reduced intensity is allowed to vary. Generally SK3(NCAS) is fixed and the scale factors on all the other ranges are varied. Not applicable for single range data NCAS = 1.
23-24	IDX(12)<0	No plot of TI(N,NC) or B(N,NC)
25-26	IDX(13)<0	Call LDSIS and calculate distances for a crystalline topology.
27-28	IDX(14)<0	Ramp(s) = 1.0 - exp(FA.s)
	>0	Ramp(s) = FA.s
29-30	IDX(15)<0	CSIS is linearly extrapolated to zero at s = 0
	>0	TSIS is used as inner region of CSIS.
31-32	IDX(16)<0	Electron diffraction data is highlighted
33-34	IDX(17)<0	AZ,BZ,CZ and DZ are read in for overlapping exponential background in C1.
	>0	Initial background points are read in C2.
35-36	IDX(18)<0	Reduced intensity is obtained by subtracting and dividing by the background.
	>0	Background is just subtracted - Recommended.
37-38	IDX(19)<0	Punch CSIS
39-40	IDX(20)<0	Ramp(s) = -100.exp(-FA.s)

COLS.	PARAMETER	DESCRIPTION
41-42	IDX(21)<0	Plot CSIS and σ(CSIS)
43-44	IDX(22)<0	Eliminates constraints on overlapping exponential background. Used only for testing.
45-46	IDX(23)<0	Number of cycles of smoothing on TI or DSIS, with square wave window of 2xKRAVG points.
47-48	IDX(24)<0	Smooth DSIS in three ranges.
	>0	No DSIS smoothing. TI is smooth if KRAVG ≠ 0 on card 4.
49-50	IDX(25)<0	Reduced intensity is obtained by subtracting a background composed of a sum of Gaussians. Alternate background treatment.
51-52	IDX(26)<0	Plot of $r^{(IPOWER-1)}G(r)$ contains individual Gaussian peaks for each of the NDIST subtracted and their sum. Gaussian decomposition display.

CONTROL CARD 3 (8F10.5)

1-10	SK	scale factor $I_{obs}/SK = I$
11-20	SZ	bulk density. This bulk density is equal to $\rho_o'(\sum_{uc} f_{s=0})^2/\sum_{uc} f_{s=0}^2$ where ρ_o' is the bulk density in units of composition per A^3.
21-30	DAMP	Experimental damping factor $\exp(-DAMP.s^2)$
31-40	FA	Ramping factor to make intensities more amenable to fitting with exponentials. Necessary for neutron data.
41-50	SKR	If SKR>0 TI(N,NC) and SKRxTI(N,NC) are plotted. Helpful when TI(N,NC) and B(N,NC) are nearly equal.
51-60	DAMA	$\alpha = -DAMA.r^2$. This is used only in plotting the final $r^{(IPOWER-1)}G(r)$ curve to reduce

		the effect of random errors at large s one feature of the radial distribution curve at large r, which arise primarily from the low angle data. DAMA is read as a positive quantity. If DAMA = 0.0 α = DAMP; if DAMA $\neq$ 0 α = -DAMP.r^2 + DAMP
61-70	R_{MAX}	Maximum value of r in the $4\pi r^{IPOWER}(\rho(r) - \rho_o)$ plot.

CONTROL CARD 4 (6I5)

1-5	NCAS	Number of overlapping data sets read in NCAS = 1 in present version of program.
6-10	NSIS2	Number of intensity data included in refinement. NSIS2 = (SLST(NC) - s(1)/ΔS + 1)
11-15	KRAVG	Running average parameter. Window is (2 x KRAVG +1) points.
16-20	IPOWER	power of r plotted in $4\pi r^{IPOWER}(\rho(r) - \rho_o)$ plot.

B. Intensity and theory section (One of the following two is required).

1. Electron diffraction input

If MTITL(20)= EDIL	Process electron diffraction data.
If MTITL(18)= DF43	Scattering factors are read from file 43 instead of cards.

1a. *Electron diffraction scattering factor coefficients.*

The electron diffraction scattering coefficients for each atom pair C_{AB}, EQ. 5, are generated in a separate program. Our program EDCOEFF is not included in RADILS. The following terms all apply to that program. The user must generate a file for these coefficients. This approach is used because the electron diffraction scattering factors are tabulated at specific s values and all of the calculations to generate C_{AB} need not be repeated each time RADILS is run.

(2I5,4X,Z4,4X,2A4)

1-5	NCOEFF	number of scattering factor coefficients
6-10	NSIS2T	number of scattering factor coefficient points.
11-14	SKIP	
15-18	Name 1	Optional name of job which created scattering factor coefficient file.
19-22	SKIP	
23-26	NAME 2	Optional date of job which created
27-30	NAME 3	scattering factor file.

(20A4) (I = 1,NCOEFF Read the next series of cards including the END card NCOEFF times.

1-4	NAME 1	First part of atom pair coefficient name eg Si
5-8	NAME 2	dash(-)
9-12	NAME 3	Second part of atom pair coefficient name e.g. O. Complete name is Si - O

(8F10.5)

1-10	DUM1	not used
11-20	TYPE	code for type of sharpening function used.
21-30	WAVENO	wavenumber $2k$ or $4\pi/\lambda$
31-40	SMN	minimum s value
41-50	SMX	maximum s value
51-60	FACTOR	not used

(2F10.5,I5)

1-10	s(1)	first s value
11-20	DS	s interval
21-25	NSIS2T	number of points

(5E14.7)

FBAR(J,I),J = 1,NSIS2T - scattering factor coefficients for the I^{th} atom pair

(20A4)

1-4	LEND	END card

Read in electron diffraction sharpening factor

(20A4)		
1-80	NTITL	"SHARPENING FACTOR" title
(8F10.5)		
1-80	DUM1,TYPE,WAVENO,SMN,SMX,FACTOR	Same as for scattering factor coefficient
(2F10.5,I5)		
1-25	s(1),DS,NSIS2T	
(5E14.7)		
	(FC(I),I=1, NSIS2T)	Sharpening function
(20A4)		
1-4	LEND	end card.

Read in electron diffraction total atomic intensity

(20A4)		
1-80	NTITL	"TOTAL ATOMIC INTENSITY" title
(8F10.5)		
1-80	DUM1,TYPE,WAVENO,SMN,SMX,FACTOR	Same as for
(2F10.5,I5)		scattering factor
1-25	s(1), DS, NSIS2T	coefficients
(5E14.7)		
	(TIA(N),N=1,NSIS2T)	Total sharpened atomic intensity
(20A4)		
1-4	LEND	END card.

1b. Electron diffraction leveled intensity if MTITL(19) = DF42 the intensity is read from file 42 instead of cards. This intensity is the output from our leveling and averaging program GETIL. RADILS expects intensity data that has been corrected for systematic errors and sharpened. If NCAS >1, read this set of cards, including END card, NCAS times. The present version has only been tested for NCAS = 1 single range data.

(20A4)	
1-8	Title card

(8F10.5)		
1-10	CH	Camera height in mm
11-20	PTOS	plate to rotating sector separation
21-30	WAVEND	wavenumber $4\pi/\lambda$
31-40	SFRST(NC)	first data print for the NC^{th} data range
41-50	SLST(NC)	last data point in the NC^{th} data range
51-60	FACTOR	not used
61-70	SF(NC)	(SK/SK3(NC)) necessary to bring the theoretical atomic intensity to the level of the NC^{th} data set for subtraction in EDIL. If SF(NC) = 0.0, no atomic theory is subtracted.
71-80	PCTSG	When standard deviations from counting statistics are not available as in electron diffraction photographic data the error in the intensity is expressed as percent error envelope. The error is expressed as a percent of the sharpening function. PCTSG = 0.005 corresponds to 0.5% of the sharpening function.
(2F10.5,I5)		
1-10	s(1)	first value of s
11-20	DS	s interval
21-25	NSIS1	number of points
(5E14.7)		
	(TI(N,NC),I=1, NSIS1)	Total intensity data for the NC^{th} data range
(20A4)		
1-4	LEND	END card

2. X-ray and neutron diffraction input

If MTITL(20)= XRT1 Process x-ray or neutron diffraction data.

If MTITL(19)= DF42 Read data from file 42.

2a. Experimental intensities.

If multiple overlapping sets of data are being processed (NCAS > 1) read the following set of cards NCAS times. Present version tested for NCAS = 1 only.

(16I5)		
1-5	NSIS1	number of total intensity data points
(8F10.5)		
1-10	s(1)	first value of s
11-20	DS	s interval
21-30	SFRST(NC)	first data point in the NC^{th} set of data
31-40	SLST(NC)	last data point in the NC^{th} set of data
41-50	SCTI	scale factor modifying TI(N,NC) and SG(N,NC) before refinement. Usually set to 1.0
(20A4)		
1-80	NFMT	variable input format for total intensities eg ((4(2F10.5)))
(20A4)		
1-80	NTITL	Title card
(NFMT)		
	(TI(N,NC),SG(N,NC),N=1,NSIS1)	Total intensity and standard deviation of the NCth data set

2b. Scattering factors

If MTITL(18) = DF43 read theory from file 43 instead of cards.

(9F8.4,F5.2) M=1, NDA		Read one card for each of the NDA distinct atom types.
1-8	AA(1,M)	The AA, BB and CC's are
9-16	BB(1,M)	the coefficients

17-24	AA(2,M)	for analytic approximations
25-32	BB(2,M)	of Hartree-Fock x-ray
33-40	AA(3,M)	scattering factors. Values are
41-48	BB(3,M)	tabulated by B. Mann, (Acta
49-56	AA(4,M)	Cryst. (1968), A24, 32))
57-64	BB(4,M)	
65-72	CC(M)	
73-77	DD(M)	number of atoms in unit of composition
78-80	card labeling information, not read	

Note 1 - For neutron diffraction data AA, BB and CC are zero and DD(M) is the coherent neutron scattering amplitude tabulated by G. E. Bacon (Acta Cryst. (1972) A28, 357).

Note 2 - The pair type indicator on the short distance parameter card NP(L) is determined by the order of the scattering factor cards. For example if the scattering factor for Si is first, followed by the O card for SiO_2

NP(L)	*DISTANCE*
1	Si - Si
2	Si - O
3	O - O

C. Initial Background Input (1 of the following three sections is required).

1. If IDX(17) < 0 read Overlapping Exponential Parameters. The background is calculated directly from the expression given in EQ. 8 Present version is only valid for NCAS = 1.

(I10,4F10.4)

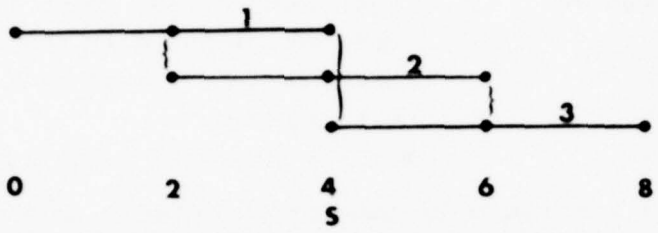
	NC=1,NCAS	If NCAS = 1 read NCAS cards
1-10	NCV(NC)	number of background segments in the NC^{th} range of overlapping data
11-20	SK5(NC)	Overall background scale factor for the NC^{th} range of overlapping data
21-30	SK3(NC)	scale factor scaling experiment to theory for the NC^{th} range of data. Usually set to 1.0 and refine SK.

31-40	SFRST(NC)	First experimental point for the NC^{th} data set.
41-50	SLST(NC)	last experimental point for the NC^{th} data set included in the refinement, NSIS2 on card 4 must be consistent with SLST(NC) $NSIS2 = (SLST(NC) - s(1)/DS + 1)$

NCV(NC) are the number of background segments in the NC^{th} background. If NCAS sets of overlapping data are being refined there will be NCAS sets of NCV(NC) background segment cards. This version is tested for NCAS = 1 only.

(4E14.7)	NC=1, NCAS	
1-14	AZ(1,NC)	Overlapping exponential
15-28	BZ(1,NC)	background segments
29-32	CZ(1,NC)	for the NC^{th} background.
33-46	DZ(1,NC)	If multiple overlapping data are
.		refined read this set for
.		each range. Present version
.		NC = 1 only.
1-14	AZ(NCV(NC),NC)	
15-28	BZ(NCV(NC),NC)	
29-32	CZ(NCV(NC),NC)	
33-46	DZ(NCV(NC),NC)	
(16F5.2)	NC=1, NCAS	
1-5	SNN(1,NC)	These are the points at which the
6-10	SNN(2,NC)	overlapping exponential background
11-15	SNN(3,NC)	segments are constrained to be
.		equal with Lagrangian
.		multipliers. The points are
.	SNN(NCV(NC),NC)	generally set equal to the
		midpoints of the background
		segments of length Δs. Read one card
		for each overlapping set of data.
		Present version NCAS = 1 only.

Example - for 3 overlapping segments of length $\Delta s = 4$



SNN(1,1) = 2.0 segments 1 and 2 are constrained equal.
SNN(2,1) = 4.0 segments 1 and 3 are constrained equal.
SNN(3,1) = 6.0 segments 2 and 3 are constrained equal.

2. IDX(17) $\geq$ 0. Read overlapping exponential trial points
This is the recommended background starting point.
Present version only tested for NCAS = 1.

(I10,7F10.4)	NC=1,NCAS	If NCAS > 1 read NCAS cards
1-10	NCV(NC)	number of background segments in the NC^{th} overlapping set of data.
11-20	SK5(NC)	overall background scale factor for the NC^{th} overlapping set of data.
21-30	SK3(NC)	scale factor placing the NC^{th} reduced intensity on an absolute scale. Usually set to 1.0 and SK refined.
31-40	SFRST(NC)	first experimental point in the NC^{th} data set.
41-50	SLST(NC)	last experimental point in the NC^{th} data set. NSIS2 on card 4 equals (SLST(NC) - s(1))/DS + 1).
(8F10.4)	NC=1,NCAS	If NCAS > 1 read NCAS cards
1-10	SS(1,NC)	SS(N,NC) and FF(N,NC) are the
11-20	FF(1,NC)	s value and trial background value,
21-30	SS(2,NC)	respectively, for the NC^{th} overlapping
31-40	FF(2,NC)	set of data. The background parameters
.	.	are determined by a three point fit

.	.	of successive trial points
.	.	eg 123; 234; 345 et.
.	SS(NCV(NC)+2,NC)	
.	FF(NCV(NC)+2,NC)	

Note 1. The selection of the SS(N,NC) are based on the segment length Δs which should be $\geq 2\pi/r^{1st}$. r^{1st} is the first bonded distance expected in sample. This is important early in the analysis to prevent background errors from interfering with the structural parameters. Δs need not be constant and in fact one or two segments $< 2\pi/r^{1st}$ in the inner region (less than 4s) can be helpful in providing the curvature needed to fit the experimental background. $\Delta s(N) = SS(N+2,NC) - SS(N,NC)$

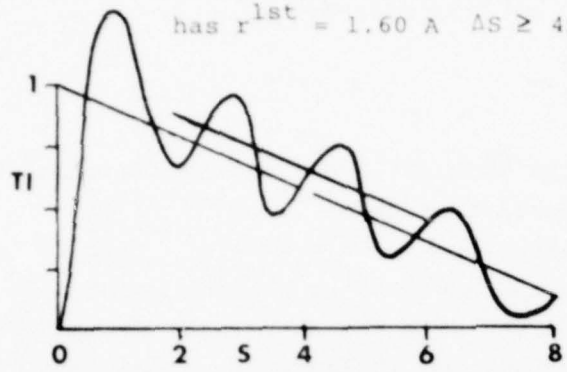
Note 2. If FF(1,NC) is zero then the initial background is determined automatically from a fit to the theoretical Σf^2 which is ramped and scaled to generate an si(s) which has equal positive and negative area. This is accomplished by minimizing the following relation

$$\sum_{N=1}^{NSIS1} s(N)/FC(N)\,[TI(N,NC)-(C_1*FC(N) + C_2 + C_3*s(N))]^2$$

this starting point may be helpful for x-ray and neutron data

(16F5.2)	NC=1,NCAS	If NCAS > 1 read NCAS cards
1-5	SNN(1,NC)	These are the points where
6-10	SNN(2,NC)	the overlapping exponential
11-15	.	segments are constrained
16-20	.	to be equal. Example
.	.	in C1 and below.
.	SNN(NCV(NC),NC)	

Note 3: The following example may help in determining Δs, SS(N,NC),FF(N,NC) and SNN(N,NC). If the sample has r^{1st} = 1.60 A $\Delta S \geq 4S$



N	SS(N,NC)	FF(N,NC)	SNN(N,NC)
1	0.05	1.0	2.0
2	2.0	0.88	4.0
3	4.0	0.58	6.0
4	6.0	0.50	
5	8.0	0.15	

3. IDX(25) < 0. Read Gaussian Summation Parameters

These initial parameters are determined from a separate program GAUSBK by fitting the Σf^2. This type of background treatment applies only to single range data NCAS = 1. IDX(14) must be > 0 so that RAMP(s) = FA.s The background is determined by the expression given in EQ. 9.

(I5,5X,3F10.5)		
5	NG	number of gaussians
6-10	skip	
11-20	FA	ramp parameter
21-30	CO	constant
31-40	SK5(1)	overall background scale factor
(8F10.5)		AX(N,1) and BZ(N,1) are the
1-10	AX(1,1)	gaussian amplitude and
11-20	BZ(1,1)	breath parameters, respect-
21-30	AZ(2,1)	ively.
31-40	BZ(2,1)	
.		
.		
.		
.	AZ(NG,1)	
.	BZ(NG,1)	

D. IDX(3) < 0 BROAD input (optional)

Used to broaden a polycrystalline pattern

(8F10.5)

1-10	PSZ	radius of the ordered region.
11-20	SLOPE	slope of a linear background which is subtracted from total intensity before broadening.
21-30	XLAM	wavelength
31-40	BINT	intercept of linear background

E. IDX(13) < 0 LDSIS input (optional)

Generates distances, coordination numbers and thermal parameters for a crystalline topology.

(9I5)

1-5	NATTY	number of distinct atom types	
6-10	NATAS	number of atoms in the asymmetric unit	
11-15	NSYM	number of symmetry transformation cards	
16-20	NX1	10-DISMX/AA	defines the number of unit cells to search in x direction for contributing atoms
21-25	NX2	10+DISMX/AA	
26-30	NY1	10-DISMX/BB	defines the number of unit cells to search in the y direction for contributing atoms
31-35	NY2	10+DISMX/BB	
36-40	NZ1	10-DISMX/CC	defines the number of unit cells to search in the z direction for contributing atoms
41-45	NZ2	10+DISMX/CC	

(8F10.5)

1-10	DISMN	Minimum distance of calculation
11-20	DISMX	Maximum distance of calculation
21-30	AA	these are the unit cells dimensions and
31-40	BB	direction cosines.
41-50	CC	
51-60	CA	
61-70	CB	
71-80	CG	

(3F10.5,I10,F10.5)		Read one card for each atom in asymmetric unit N=1,NATAS.
1-10	XX(N)	Fractional coordinates of the N^{th} atom
11-20	YY(N)	in the asymmetric unit.
21-30	ZZ(N)	
31-40	NNTYPE(N)	atom type
41-50	WT(N)	Weight used to generate proper coordination number. The weight can be set to be the reciprocal of the square root of the number of atoms of the 1^{st} type, NTYPE(1), in the asymmetric unit.
(12F5.2)		Read one card for each symmetry operation. NSYM cards
1-5	DX(N)	DX(N) DY(N) DZ(N) is the translational vector for the N^{th} symmetry operation
6-10	FX1(N)	
11-15	FX2(N)	
16-20	FX3(N)	
21-25	DY(N)	FX1(N) FX2(N) FX3(N) FY1(N) FY2(N) FY3(N) FZ1(N) FZ2(N) FZ3(N) is the rotational symmetry transformation for the N^{th} symmetry operation. See symmetry transformation in Glossary.
26-30	FY1(N)	
31-35	FY2(N)	
36-40	FY3(N)	
41-45	DZ(N)	
46-50	FZ1(N)	
51-55	FZ2(N)	
56-60	FZ3(N)	
(8F10.5)		
1-10	RDD	value of the first bonded distance
11-20	WDD	amplitude for first bonded distance
21-30	RDD2	distance and amplitude of any other
31-40	WDD2	distance for which a good approximation of the amplitude is known.
41-50	AMPMAX	maximum value of the amplitude at large values of r.

F. Short distance parameter imput (required)

(16I5)

5	NDIST	number of short distances

(F10.6,I10,2F10.6) Read NDIST cards. One for each short distance L=1,NDIST

1-10	A(L)	Coordination number for the L^{th} distance
11-20	NP(L)	Pair type of the L^{th} distance. Determined by the position of the scattering factor cards in B1a and B2b.
21-30	XLAB	B(L) = -XLAB**2/2.0. XLAB is the root-mean square amplitude of vibration for the current distance.
31-40	RZ(L)	The L^{th} interatomic distance.

Note: If IDX(7) < 0 G(r) will be back transformed out to RZ(NDIST) to get a better approximation of the overlapping exponential background points. These points are printed after the total intensity-background plot and punched on unit 41. They can be used as starting points when resubmitting the job.

G. Refinement section (required)

The KI(N) vector specifies which parameters are refined and the type of background treatment. There are a total of NPAR flags. NPAR = 3*NDIST + 4. The first two flags refer to scale factor and bulk density, the next 3*NDIST refer to the structural parameters of the NDIST short distances and the last two refer to the type of background treatment and the scaling of the observed data. Generally the last flag is set to 1 and SK the scaling of theory to experiment is varied. The background flag is NPARM = NPAR -1 and the structural parameter flags end at NNP = NPAR - 2.

(5I2,35I2,(140I2))

1-2	ID1	ID1 = 1 eigenvector-eigenvalue procedure is used to obtain parameter shifts.

		ID1 = 0 matrix inversion is used
3-4	NCON	number of imposed constraints on structural parameters. Background constraints are applied automatically in program using Lagrangian multipliers.
5-6	ID3	not used.
7-8	NADD	NADD = 1 Gaussian description of short distance peaks are reintroduced into the $4\pi r^{IPOWER}(\rho(r) - \rho_o)$ plot. NADD = 0 short distances are not reintroduced.
9-10	NSUB	NSUB = 1 Plots of TI(N,NC), B(N,NC), SIS(N,NC) CSIS(N), σ(CSIS(N)), G(r), $4\pi r^{IPOWER}(\rho(r) - \rho_o)$ are displayed before current refinement cycle.
11-12	KI(1)	KI(1) = 2 SK scale factor is refined. KI(1) = 1 SK scale factor scaling theory to to experiment is not refined.
13-14	KI(2)	KI(2) = 3 SZ bulk density is refined. KI(2) = 1 SZ bulk density is not refined.
15-16	KI(3)	KI(3) = 4 Coordination number of first short distance is refined. KI(3) = 24 Coordination number of first short distance is constrained. Q matrix will specify which other coordination numbers are involved in the constraint relation. KI(3) = 1 Coordination number is not refined.
17-18	KI(4)	KI(4) = 5 The first short distance is refined. KI(4) = 25 The first short distance is constrained. Q matrix will specify which other distances are involved in the constraint relation. KI(4) = 1 The first short distance is not refined.

19-20	KI(5)	KI(5) = 6 Thermal parameter for the first short distance is refined.
		KI(5) = 26 Thermal parameter for the first short distance is constrained. The Q matrix will specify which other thermal parameters are involved in constraint relation.
21-22	KI(6)	Coordination number flag for second short distance
23-24	KI(7)	Second short distance flag.
25-26	KI(8)	Thermal parameter flag of second short distance.
.	.	The same numbering scheme as in KI(3), KI(4),
.	.	and KI(5) is used.
.	.	
.	KI(NNP)	Thermal parameter of the NDIST distance
.	KI(NPARM)	KI(NPARM) = 7 A and B of each overlapping exponential background segment is refined.
		KI(NPARM) = 8 A and C of each overlapping exponential background segment is refined.
		KI(NPARM) = 9 SK5 overall scale factor of overlapping exponential background is refined.
		KI(NPARM) = 10 A,B and C of each overlapping exponential background segment is refined.

	KI(NPARM) = 12	A,B and D of each overlapping exponential background segment is refined.
	KI(NPARM) = 13	SK5, CO, FA of Gaussian summation background is refined.
	KI(NPARM) = 14	A and B of each gaussian is refined.
KI(NPAR)	KI(NPAR) = 11	SK3 scaling of experiment to theory is refined. Usually set to 1.0 and not refined.
	KI(NPAR)= 1	SK3 not refined.

If NCON > 0 Read Q(N,M) matrix which specifies which of the original NPARR parameters read on the KI card are to be varied independently (upper section NIV x NPARR) and how the dependent parameters are related (lower section NCON x NPARR). The upper section is automatically determined in the program from the KI card i.e. all flags less than $\leq$ 20. The lower section is read from NCON data cards. These cards represent the last NCON rows of Q. Each row is composed of the coefficients of all parameters involved in the linear constraint relation. Read one card for each constraint relation. NCON cards.

(10F8.3) L = NPARR - NCON + 1

1-8	Q(L,1)	If these parameters are involved in the
9-16	Q(L,2)	constraint relation, these are the
17-24	Q(L,3)	coefficients.

.
.
.

EXAMPLE OF KI CARD

Type of Refinement	N	KI(N)	Parameter Refined
Structural Parameters Eq. 9 Parameters can be constrained.	1	2	SK (scaling of theory to experiment)
	2	3	SZ
	3	4	N1
	4	5	r1
	5	6	l1
	6	24**	N2
	7	5	r1
	8	6	l2
	9	1	not refined N(NDIST)
	10	5	r(NDIST)
	NNP	6	l(NDIST)
Overlapping Exponential Background Eq. 10	NPARM*	7	A,B (Suggested parameters)
		8	A,C
		9	SK5
		10	A,B,C
		12	A,B,D
Gaussian summation Background Eq. 11		13	SK,C0,FA
		14	A,B
	NPAR	11	SK3 (scaling of experiment to theory)

* CHOOSE ONLY ONE. If KI(NPARM) = 1 skip background refinement.

** The second coordination number is constrained. Q matrix specifies which other coordination numbers are involved in constraint relation.

Q(N,NPARR)

example KI card 2 3 4 1 1 24 1 1 24 1 1 9 1

NCON = 2 NIV = 4 NPARR = 6

$1.5N_1 - N_2 = 0$ and $0.5N_1 - N_3 = 0$

Q

		SK	SZ	N1	N2	N3	SK5			
U	1	1.0						SK		SK
			1.0					SZ		SZ
				1.0				N1		N1
	NIV						1.0	N2	=	SK5
				1.5	-1.0			N3		0
L	NPARR			0.5		-1.0		SK5		0

The upper section U automatically sets Q(1,1), Q(2,2), Q(3,3) and Q(4,6) to 1.0. N_2 and N_3 were defined as 24, instead of 4, so they are treated as dependent variables.

The lower section L is read from two cards.

1-8	Q(5,1)	0.0
9-16	Q(5,2)	0.0
17-24	Q(5,3)	1.5
25-32	Q(5,4)	-1.0
33-40	Q(5,5)	0.0
41-48	Q(5,6)	0.0

1-8	Q(6,1)	0.0
9-16	Q(6,2)	0.0
17-24	Q(6,3)	0.5
25-32	Q(6,4)	0.0
33-40	Q(6,5)	-1.0
41-48	Q(6,6)	0.0

(F5.2,2(F5.2,I5,2F5.2),3F5.2)

1-5	SFT	Applied shifts are SFT times calculated shifts
6-10	FOBS(1)	first r value in the first observational range of the least squares. This is the inner region of $G'(r) + 4\pi r\rho_o$ $(4\pi r\rho(r)$.
11-15	NOBS(1)	Number of observations in the inner region of $4\pi r\rho(r)$
16-20	DOBS(1)	increment in r in the inner region of $4\pi r\rho(r)$
21-25	WOBS(1)	weight of the inner region of $4\pi r\rho(r)$ in the refinement.
26-30	FOBS(2)	First r value in the second observational range of the least squares. This is the outer region of G'(r).
31-35	NOBS(2)	Number of observations in outer region of G'(r). Set to 1 if just refining inner region.
36-40	DOBS(2)	increment in r in the outer region of G'(r).
41-45	WOBS(2)	weight of the outer region of G'(r) in the refinement.
46-50	WOBS(3)	weight of the third observational range.
51-55	WOBS(4)	weight of the fourth observational range.
56-60	WOBS(5)	weight of the fifth observational range. These last three observational ranges are used when overlapping sets of data are being refined. They are the three possible overlap regions in the reduced intensity from a maximum of four overlapping sets of total intensity data. The reduced intensity CSIS(N) must be continuous and therefore the two intensity contributors in an overlap region must be restricted This can be done with constraints on the end points or by restraining their difference to be below a certain limit. These weights are used when the overlap regions are restrained. The constraints are selected with the IDX control card. If constraining these weights are 0.

If ID1 > 0

(3F10.5,I10)

1-10	EVMIN	minimum eigenvalue accepted in filtering process in IGN. If 0 all eigenvalues are accepted.
11-20	RSMIN	minimum residual reduction required in filtering process in IGN for a particular eigenshift.
21-30	SHIFT	maximum eigenshift (linear parameter shift) allowed in filtering process in IGN.
31-40	MAXPAR	maximum number of eigenvalues (linear parameters) allowed in filtering process in IGN. Program selects the MAXPAR eigenshifts which produce the greatest residual reduction.

IV. OPERATIONAL REMARKS

A. Constraints

The interatomic distances (RZ(L)), the coordination numbers (A(L)), and the disorder parameter (B(L)) can be constrained by NCON linear constraint relations, Q, which are read in RADILS. If NCON = 0 no constraints are used. The imposition of constraints lessens the number of variables (NPARR) and required suitable combinations of the original derivatives in constructing a new derivative matrix for the smaller set of independent variables, NIV. The new derivatives, with respect to the independent parameters, v, can be obtained from the old derivatives, with respect to the original dependent parameters, x, by

$$\left(\frac{dF}{d\underline{v}}\right) = J^T \left(\frac{dF}{d\underline{x}}\right)$$

where F is the function being minimized, T is the transpose operation and $\underline{J} = \underline{Q}^{-1}$. The original parameter changes are obtained in RESET from the independent set by $\Delta\underline{x} = \underline{J}\,\Delta\underline{v}$.

The Q matrix consists of an upper section $\underline{U}$ (NIV x NPARR) and a lower section $\underline{L}$ (NCON x NPARR). The upper section determines which of the original parameters will be treated as linearly independent variables and the lower section summarizes all the constraints imposed on the problem, by specifying the relationships between the dependent and independent variables.

$$\begin{vmatrix} \underline{U} \\ \underline{L} \end{vmatrix} d\underline{x} = \begin{vmatrix} d\underline{v} \\ 0 \end{vmatrix}$$

$$Q\, d\underline{x} = d\underline{v}$$

The upper section is automatically calculated from the KI card. When a structural parameter flag 4, 5 or 6 is entered as 24, 25 or 26 that parameter is considered to be constrained. $\underline{U}$ is found by placing ones and zeros in appropriate locations.

The lower section L is read on the cards following the KI card if NCON > 0. The coefficients of the linear constraint relations are placed by the user in the appropriate column. The record length for each constraint relation is NPARR x 8 fields. If NPARR is > 10 more than one card will be required.

Example: Silica glass - We would like to vary the scale factor SK, bulk density SZ, the coordination numbers of the Si-O, O...O and Si...Si and the overall background scale factor SK5.

In addition we would like to constrain the coordination numbers as follows

$$1.5 \times N^{1}_{Si\text{-}O} \; -- \; N^{2}_{O\ldots O} = 0 \qquad N^{1} = N_{Si\text{-}O}$$

$$0.5 \times N^{1}_{Si\text{-}O} \; -- \; N^{3}_{Si\ldots Si} = 0 \qquad N^{2} = N_{O\ldots O}$$

$$N^{3} = N_{Si\ldots Si}$$

B. Solution of Normal Equation

The user has a choice in obtaining the shift vector. Either INVERT or IGN can be used depending on the complexity of the problem. If ID1=1 IGN is selected and the shifts are determined by the eigenvalue-eigenvector technique. Several limiting parameters, read in RADILS, can help with unstable matrices by filtering out eigenvalues which are either too small, cause excessive shifts in the linear combination parameters or insufficient reduction in the linear approximation residual. Since the eigenshifts are uncorrelated, setting any of them to zero, if it does not meet a particular criterion, does not affect the required values of the remaining ones. Thus filtering provides a means of ensuring that large eigenshifts will not be allowed unless the resultant residual reduction is judged sufficient. It is even possible to refine correlated parameters simultaneously, using IGN i.e. SK, SZ, N, R1, SK5.

Invert uses the ASC system matrix inversion DSIMEC, which is a double precision routine operating on a linear packed array APACK. SIMEC (a single precision routine) and USIMEC which inverts a matrix which is a different order than the dimension statement, are also used throughout RADILS. Appropriate system calls will have to be applied.

C. Interference Intensity Calculation

I. By substracting a background scattering curve

$$si(s) = (I_t(s) - B(s))/\Sigma f^2$$

Background line is calculated from

A. Series of overlapping exponential curves

1. Starting parameters obtained by a guess at the background scattering of selected values of s.
2. Starting parameters obtained from selected values of Σf^2, which has been scaled to the experimental total intensity.

B. Summation of Gaussian curves

1. Starting parameters are determined in a separate program by a trial and error fit to the Σf^2.

II. By subtracting and dividing by a background curve. The background is calculated as in C1 and 2 only.

$$si(s) = I_t(s)/B(s) + 1.0$$

D. A few Texas Instrument - ASC system routines are used for speed and convenience. These system routines must be replaced. Enclosed is a description of the name and uses of the ASC routines used to facilitate substitution.

SIMUL EQUATIONS / MATRIX INVERSION / DETERMINANT

1. ENTRY POINTS

MATRICES STORED BY COLUMNS:

CALL SIMEC (A,B,N,M,D,I)	REAL*4	ARGUMENTS
CALL DSIMEC(A,B,N,M,D,I)	REAL*8	ARGUMENTS
CALL CSIMEC(A,B,N,M,D,I)	C*8	ARGUMENTS
CALL CDSIMC(A,B,N,M,D,I)	C*16	ARGUMENTS

MATRICES STORED BY ROWS:

CALL SIMER (A,B,N,M,D,I)	REAL*4	ARGUMENTS
CALL DSIMER (A,B,N,M,D,I)	REAL*8	ARGUMENTS
CALL CSIMER (A,B,N,M,D,I)	C*8	ARGUMENTS
CALL CDSIMR (A,B,N,M,D,I)	C*16	ARGUMENTS

GENERALIZED SIMULTANEOUS EQUATIONS:

MATRICES STORED BY COLUMNS

CALL LSIMEC(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	REAL*4	ARG
CALL DLSIMC(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	REAL*8	ARG
CALL CLSIMC(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	COMPLEX*8	ARG
CALL CDLSMC(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	COMPLEX*16	ARG

MATRICES STORED BY ROWS

CALL LSIMER(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	REAL*4	ARG
CALL DLSIMR(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	REAL*8	ARG
CALL CLSIMR(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	COMPLEX*8	ARG
CALL CDLSMR(X,Y,K,L,D,IERAS,IX,JX,IY,JY)	COMPLEX*16	ARG

2. PURPOSE

TO SOLVE SEVERAL SETS OF SIMULTANEOUS EQUATIONS, INVERT A MATRIX, OR BOTH. IT CAN ALSO BE USED TO EVALUATE A DETERMINANT.

3. ARGUMENT DEFINITIONS

INPUT

A MATRIX OF COEFFICIENTS
B MATRIX OF CONSTANTS
N NUMBER OF ROWS
M NUMBER OF COLUMNS IN B (MAY BE ZERO)
I BLOCK OF ERASABLE STORAGE AT LEAST 4N IN LENGTH.

OUTPUT

A INVERTED MATRIX

SIMUL EQUATIONS / MATRIX INVERSION / DETERMINANT - CONTINUED

B MATRIX OF ROOTS
D DETERMINANT

GENERALIZED SIMULTANEOUS EQUATIONS:

X INPUT MATRIX OF COEFFICIENTS DIMENSIONED (IX,JX)
Y VECTOR OR MATRIX OF CONSTANTS DIMENSIONED(IY,JY)
N NUMBER OF ROWS AND COLS OF "X" TO BE USED.
L NUMBER OF ROWS(USIMEC) OR COLS(USIMER) OF Y TO USE.
D DETERMINANT (OUTPUT)
IERAS BLOCK OF ERASABLE STORAGE (3K IN LENGTH)
IX ROW DIMENSION OF "X"
JX COL DIMENSION OF "X"
IY ROW DIMENSION OF "Y"
JY COL DIMENSION OF "Y"

4. METHOD

IF ONLY MATRIX INVERSION OR DETERMINANT EVALUATION IS DESIRED, SET "M" TO ZERO. HOWEVER, "B" MUST STILL BE PRESENT.
ALTHOUGH IT IS ONLY A DUMMY.

5. EXAMPLES

USE SIMEC:

```
 A(1,1)  A(1,2)  ...   A(1,N)
 A(2,1)  A(2,2)  ...   A(2,N)
          ...
 A(N,1)  A(N,2)        A(N,N)

 B(1,1)  B(1,2)        B(1,M)
 B(2,1)  B(2,2)        B(2,M)
          ...
 B(N,1)  B(N,2)        B(N,M)
```

CALL SIMEC(A,B,N,M,D,I)

SOLVES

$$AX = \begin{vmatrix} B(1,1) \\ B(2,1) \\ \vdots \\ B(N,1) \end{vmatrix}, \quad AX = \begin{vmatrix} B(1,2) \\ B(2,2) \\ \vdots \\ B(N,2) \end{vmatrix} \quad \ldots \quad AX = \begin{vmatrix} B(1,N) \\ B(2,N) \\ \vdots \\ B(N,M) \end{vmatrix}$$

6. RESTRICTIONS

THE MATRICES "A" AND "B" ARE DESTROYED BY EXECUTION OF THIS SUBROUTINE.

SIMUL EQUATIONS / MATRIX INVERSION / DETERMINANT - CONTINUED

"A", "B", AND "D" ARE REAL*4, REAL*8, COMPLEX*8 OR COMPLEX*16 DEPENDING ON THE "CALL" STATEMENT.

WARNING
THE MATRICES "A" AND "B" MUST BE PACKED; THAT IS, "A" MUST MUST BE DIMENSIONED (N,N) AND "B" MUST BE DIMENSIONED (N,M) OR (M,N) DEPENDING ON WHETHER THE MATRICES ARE STORED BY ROWS OR COLUMNS. FOR EXAMPLE, IF IT IS DESIRED TO INVERT

$$A = \begin{vmatrix} 1 & 3 \\ 2 & 4 \end{vmatrix}$$

AND "A" IS INCORRECTLY DIMENSIONED (3,3), THEN "A" WILL CONTAIN THE 9 NUMBERS:

1,2,X,3,4,X,X,X,X

AND THE SUBROUTINE "SIMEC" WILL ATTEMPT TO INVERT THE MATRIX

$$\begin{vmatrix} 1 & X \\ 2 & X \end{vmatrix}$$

MATRIX MULTIPLICATION

1. ENTRY POINTS

```
CALL MMPY  (I,J,K,A,B,C)              REAL*4      ARGUMENTS
CALL DMMPY (I,J,K,A,B,C)              REAL*8      ARGUMENTS
CALL CMMPY (I,J,K,A,B,C)              COMPLEX*8   ARGUMENTS
CALL CDMMPY(I,J,K,A,B,C)              COMPLEX*16  ARGUMENTS

GENERALIZED MATRIX-MATRIX MULTIPLICATION:
CALL   LMMPY(L,M,N,X,Y,Z,IX,JX,IY,JY,IZ,JZ)  REAL*4      ARG
CALL  DLMMPY(L,M,N,X,Y,Z,IX,JX,IY,JY,IZ,JZ)  REAL*8      ARG
CALL  CLMMPY(L,M,N,X,Y,Z,IX,JX,IY,JY,IZ,JZ)  COMPLEX*8   ARG
CALL  CDLMMY(L,M,N,X,Y,Z,IX,JX,IY,JY,IZ,JZ)  COMPLEX*16  ARG
```

2. PURPOSE

TO MULTIPLY TWO MATRICES TOGETHER

3. ARGUMENT DEFINITIONS

```
I   NUMBER OF ROWS IN "A" AND "C".
J   NUMBER OF COLS IN "A" AND NUMBER OF ROWS IN "B".
K   NUMBER OF COLS IN "B" AND "C"
A   INPUT MATRIX   (LEFT MULTIPLIER)
B   INPUT MATRIX   (RIGHT MULTIPLIER)
C   OUTPUT MATRIX

GENERALIZED MATRIX-MATRIX MULTIPLICATION:

L   ROW DIMENSION OF "X" AND "Z" TO BE USED
M   COL DIMENSION OF "X" AND ROW DIMENSION OF "Y" TO USE
N   COL DIMENSION OF "Y" AND COL DIMENSION OF "Z" TO USE
X   INPUT MATRIX DIMENSIONED (IX,JX)
Y   INPUT  MATRIX DIMENSIONED (IY,JY)
Z   OUTPUT MATRIX DIMENSIONED (IZ,JZ)
IX  ROW DIMENSION OF "X"
JX  COL DIMENSION OF "X"
IY  ROW DIMENSION OF "Y"
JY  COL DIMENSION OF "Y"
IZ  ROW DIMENSION OF "Z"
JZ  COL DIMENSION OF "Z"
```

4. METHOD

$$C(I,K) = A(I,1)*B(1,K) + A(I,2)*B(2,K) + \ldots + A(I,J)*B(J,K)$$

MATRIX MULTIPLICATION - CONTINUED

5. RESTRICTION

"C" MAY NOT BE THE SAME MATRIX AS EITHER "A" OR "B"

FOR THE GENERALIZED ROUTINES:
L <= IX,IZ
M <= JX,IY
N <= JY,JZ

V. GLOSSARY

A(L) - Coordination number for the Lth distance. Corresponds to N_{AB} in Eq. 5.

AA, BB, CC, DD - parameter for calculation of x-ray or neutron atomic form factors. Used only in XRTI.

AA, BB, CC - are the unit cell dimension in LDSIS. CA, CB, CG are the direction cosines.

AK (100,100) - is the coefficient matrix in RADILS. After solution of the problem Ax = B either by matrix inversion or the eigenvector-eigenvalue method AK contains the inverse matrix which is used to determine standard deviations and the correlation matrix. AK also serves as product matrix in IGN.

AL(100,100) - scratch array used to store coefficient matrix in INVERT.

AM(100,100) - scratch array used for product matrix in INVERT.

AMPMAX - is the largest value of the amplitude at large r.

AP(3,3), BP(3) - arrays used in CURVE to obtain initial overlapping background curve parameters from the Σf^2.

APACK(10,000),BPACK(100) - packed vectors containing AK and BK in INVERT.

AT(100,100) - is the transpose of the eigenvector matrix CK.

AZ(M,NC), BZ(M,NC), CZ(M,NC), DZ(M,NC) are the exponential shape parameters of the mth segment of the NCth background line.

B(L) - the B values of the Lth distance. Corresponds to $\frac{1}{2}1^2_{AB}$ in EQ. 5.

BAR(K,NL) - is a normalized scattering factor for atom NL at the Kth value of S. It can be expressed as

$$f_{NL}(s)/\sqrt{\sum_{uc} f^2}$$

BK(100) - is the weighted residual vector in RADILS. After matrix inversion or the eigenvector-eigenvalue method the parameter changes are stored in BK.

BL(100) - scratch array used to store residual vector BK in INVERT.

BLK(4000) - is a scratch array used in IGN and ultimately contains the linear parameter changes.

BROADN(600) - scratch array used to calculate broadened total intensity of a crystalline topology in BROAD.

CDIS(3500,10) - is the coordination number for each type of distance in bins of 0.01A out to DISMX.

CH - the camera height for the current data set. Used only in EDIL.

CK(100,100) - Coefficient matrix AK is stored in CK in IGN. After call to EIGRS the eigenvector matrix X is stored in CK.

CO, FA, SK5 - parameters used in the gaussian summation background line

$$B(s) = SK5 \left(\sum_{m=1}^{NG} A_n \exp(-B_n s^2) + FA.s + CO \right)$$

CSIS(N) - the combined reduced intensity corresponding to si(s) in EQ. 5 at s(N). For multiple data ranges the individual intensities are combined in SUBSIS.

DAMP - Damping factor applied to DSIS in SUBSIS, and used in the refinement. DAMP corresponds to α in EQ. 7

DAMA - the value of α in EQ. 7 can be set equal to $-DAMA.r^2$ for the final radial distribution plot. This variation of α with respect to r was introduced in order to diminish the effect of random errors at larger s on the features of the RDF at large r, which arise primarily from the low angle data.

DEL(L) - the Lth observed value

$$DEL(L) = \begin{cases} r\rho(r_L) & \text{for } 1 \le L < NOBS(1) \\ r\left(\rho(r_L) - \rho_o\right) & \text{for } NOBS(1)+1 \le L \le NOBS(1)+NOBS(2) \\ \Delta si(s) & NOBS(1)+NOBS(2)+1 \le L \le NOBS(1)+NOBS(2)+NOBS(3) \\ \Delta si(s) & NOBS(1)+NOBS(2)+NOBS(3)+1 \ \ L \text{ ----} \end{cases}$$

etc.

DET - the value of the determinant obtained from the ASC system matrix inversion routine.

DEUW - is the total standard deviation after the current refinement cycle.

$$DEUW = \sqrt{\sum_{I=1}^{NRHO} DEL(I)^2/(NRHO-NPARR+NCON)}$$

DEX(20) - array used in CURVE to obtain initial value of c in $\exp(a + bs^c)$ from a 3-point fit to initial background points.

DIJ(L,ICT) - derivative matrix DIJ(200,50). It represents the derivative of the ICT parameter evalued at the Lth observed value.

DIND(200,50) - is the derivative matrix of independent parameters when constraints are used on the structural parameters.

DISMN - is the minimum distance at which LDSIS calculates the longer distance (LD) contribution in LDSIS. SD refers to the short ordered distances.

$$si'(s) = si(s) - \left(si_{SD}\Big|_{0}^{NDIST} + si_{LD}(s)\Big|_{DISMN}^{DISMX}\right)$$

DISMX - is the maximum value of r where the long distance contributors are calculated in LDSIS.

DK(100) - Weighted residual vector BK is stored in DK in IGN.

DOBS(K) - Increment of N for the Kth observation ranges. Read in only for K = 1 & 2.

DS - the increment in Δs

DSIS(N) = CSIS(N) - TSIS(N). This corresponds to si'(s) in EQ. 5.

DUM(100) - scratch array used in USIMEC, ASC system matrix inversion.

DUM1 - not used.

DX(12), DY(12), DZ(12) -cell translation for x, y and z respectively on symmetry transformation card in LDSIS.

ED(27) - the array used to print out the final normalized values of the data in SUBTR.

EPS = $1.0 - 1.5 \times r/D + 0.5\ (r/D)^3$ $r \leq D$.
EPS is a spherical particle size function which corrects the relative frequency of occurrence of an interatomic vector in a structure of unbound dimension to that which pertains within a sphere of diameter D.

ESIS(600) - scratch matrix used to smooth DSIS in SUBSIS.

EV(100) - is initially the vector of eigenvalues in IGN. Subsequent to call to EIGRS the inverse eigenvalues are stored in EV.

EVMIN - minimum eigenvalue accepted in filtering process in IGN.

EVSAV(100) - the eigenvalues in IGN are stored in EVSAV.

FA - Parameter used in ramping function RAMP(S). If IDX(14)<0 RAMP(s) = 1.0 - EXP(FA.5) otherwise RAMP(S) = FA.s. If IDX(20)<0 RAMP(s) = $-100\ 1^{-FA.s}$. This function could be experimented with, to obtain the best shape for background fitting.

FACTOR - not used.

FBAR(N,NL) - is the scattering factor coefficient for the NLth atom pair at the Nth value of s. It can be expressed as

$$\frac{f_A(s) \cdot f_B(s) \cdot \cos\ (\eta_B(s) - \eta_A(s)}{I_s(s)}$$

the f and η represent the magnitude and phase of the coherent atomic scattering factor. The cos term is only used in ED.

FC(N) - is the sharpening function $I_s(s)$. It is calculated for XD and ND in XRTI from the scattering factors and it is read from file for ED. $I_s(s) = \sum_{uc} f^2$ where uc represent the unit of composition.

FF(I) - the value of the Ith point of the initial background for the current data range. Used in CURVE to generate initial background parameters.

FJ - $DS/(2\Pi^2)$

FOBS(K) - the first r-value for the Kth observation range. FOBS(K) is different from zero only for K = 1 (inner region of the RDF) and for K = 2 (outer region of the RDF).

FT(N,NC) - Continuous composite background for the NCth data set of the Nth r-value made up of weighted overlapping exponential background segments.

$$FT(NC) = \sum_m W_{km}(s) \exp\left(a_{km} + b_{km} * s^{C_{km}} + d_{km} \cdot s\right)$$

GK(100,100) - scratch array used in IGN for the eigenvectors and dimensioned in RADILS.

IA(100) - scratch array used in USIMEC the ASC system matrix inversion.

ICT - is an index of the parameters in the derivative matrix DIJ.

IDX(I) - the flag array used to control the various options.

IN(I,NC) - the integer corresponding to SNN(I,NC).

IPOWER - the power of N in the RDF plot. $4\pi r^{IPOWER} (\rho(n) - \rho_o)$ is plotted. The default value is 1.

ITYPE - Flag for treatment of the background. If IDX(18) $\geq$ 0 the empirical background is subtracted to obtain SIS. If IDX(18)<0 SIS is obtained by subtracting and dividing by the background.

KI(N) - the variation flag for the Nth parameter. Refer to refinement section for clarification.

KRAVG - Running average parameter. If greater than 0, a (2* KRAVG + 1) point running average on the total intensity is performed so that the correlation length is KRAVG*DS. If IDX(24)<0 DSIS is smooth in three stages. The data from 0.05 - 5.0 are not altered. On the first smoothing cycle ESIS is smooth with "IDX(23)" passes of a (2*KRAVG + 1) point square wave and the

data in DSIS from 5.0 to NSIS2 are replaced. On the second smoothing cycle the already smooth ESIS is again smooth as before and the data in DSIS from 15.05 to NSIS2 are replaced.

M1 - set to 100 - not used in program.

MAXPAR - maximum number of linear parameters (eigenvalues) allowed in IGN.

MTITL - Title card for job.

N1 - the number of variables + background Lagrangian multipliers. It is the order of the matrix AK, CK, AT, X.

NADD - Flag for reintroducing the short distance peaks (without termination errors or errors due to non-constant coefficients) into the plot of the RDF. NADD = 0 distances are not re-introduced. NADD = 1 distances are.

NAME2, NAME3 - is the date of the job which created the ED scattering factor file.

NAME1 - also represents the JOB ID of the job which created the ED scattering factor file.

NAME2 - dash(-)

NAME1, NAME3 - alphanumeric atom name for ED scattering factor coefficients.

NATAS - number of atoms in the asymmetric unit in LDSIS.

NATTY - number of distinct atom types in sample in LDSIS.

NC - the index which normally corresponds to the data range currently being processed.

NCAS - the number of ranges of data to be processed. The maximum number of ranges is 4. The present version of the program has been tested for NCAS = 1 only.

NCIN - flag for the type of background input. If IDK(17)< 0, NCIN = 1 and AZ's, BZ's, CZ's and DZ's for the exponentials are read in. If IDX(17) ≥ 0, NCIN = 0 and AZ's, BZ's, CZ's and DZ's are calculated from points on the initial background which are read in.

NCOEFF - the number of distinct coefficient types to be read in. Used in EDIL.

NCON - number of constraints imposed on the structural parameters in CON.

NCV(NC) - The number of background curves making up the background for the NCth data range. JK is often used to be the number of curves for the current data range.

NDA - the number of distinct atom types in the unit of composition.

NDIST - the number of short distances for which parameters will be read in.

NFMT - the format to be used for the input of experimental x-ray or neutron intensities.

NG - number of gaussians used in Gaussian summation background line.

$$B(s) = SK5 \left(\sum_{n=1}^{NG} A_n \exp\left(-B_n s^2\right) + FA.s + CO \right)$$

NIV - is the number of independent variables when the structural parameters are constrained.

NNTYPE(300) - atom type identification for atoms in asymmetric unit in LDSIS.

NOBS(K) - the number of observations in the Kth observation range.

K = 1 (inner region of $G'(r) + 4\pi r\rho_o$)

K = 2 (outer region of $G'(r)$)

K = 3,4,5 (overlap of various data sets.)

only NOBS(1) and NOBS(2) are calculated.

NP(L) - the pair type of the Lth distance.

NPAR - number of flags on KI card.

NPARM - location of flag on KI card indicating background treatment.

NPARR - the number of variables being refined.

NRHO - Total number of observations Σ NOBS(N). The derivative matrix is NRHO X NPARR.

NSD - number of ranges over which a standard deviation is calculated NSD = 2 + (NCAS -1). 2 is for the inner and outer region of the RDF and (NCAS-1) is the number of overlap overlap regions in the si(s) curve for NCAS ranges of data.

NSIS1 - the index of the largest experimental s-value.

NSIS2 - Index of the largest s-value used in the least-square refinement.

NSIS2T - number of ED scattering factor coefficient points on input file tc EDIL.

NSUB - Flag for displaying the intensities and RDF before the current refinement cycle begins. NSUB = 0 for no display and NSUB = 1 for a display.

NSYM - number of symmetry transformations in LDSIS.

NTITL(I) - Title array describing the current data set used in EDIL.

NX1 = 10 - DISMX/AA
NX2 - 10 + DISMX/AA - define the number of unit cells in the x direction in which to search for contributing atoms in LDSIS.

NY1 = 10 - DISMX/BB
NY2 = 10 + DISMX/BB - define the number of unit cells in the y direction in which to search for contributing atoms in LDSIS.

NZ1 = 10 - DISMX/CC
NZ2 - 10 + DISMX/CC define number of unit cells in the z direction to search for contributing atoms in LDSIS.

PCTSG - A constant error expressed as a percent of the sharpening function. Used in EDIL to generate an SG array. PCTSG = 0.005 corresponds to 0.5% of the sharpening function.

PER(100), PERSAV(100) - are the fractional linear residual reduction.

PLOTA - ASC system scratch array used for plotting.

PSZ - is the radius of a spherical ordered region with a particular bonding topology.

PTDS - the plate to sector separation for the current data set. Used only in EDIL.

Q(100,100) is a matrix (NPARR X NPARER) which is composed of the coefficients of the dependent parameters in the linear constraint relations. The NIV independent parameters occupy the first NIV rows of Q with ones in the appropriate columns.

QINV(100,100) - inverse of Q equal to J matrix in constraint writeup.

QT(100,100) - transpose of Q^{-1}. Described as J^t in constraint writeup and relates the dependent and independent derivatives.

RAMP(S) - A ramping function used to make the observed background more amenable to fitting with a sum of exponentials.

RAMP(S) = 1.0 - exp (FA*S) if IDX(14) < 0

or

= FA*s if IDX(14) $\geq$ 0

RDD, WDD, RDDZ, WDDZ, AMPMAX - An exponential fit of the function AMP = AMPMAX (1.0 - EXP (Ax + Bx x r) to these parameters is used in LDSIS to determine the thermal parameters for the longer distances in the intensity curve EXP ($-s^2AMP^2/2.0$).

RDD and WDD are the distance and amplitude for the first bonded distance.

RDD2 and WDD2 - are the distance and amplitude for any other distance for which a good approximation for the amplitude is known.

RMAX - the maximum value of r (in A) to be plotted in the RDF. Used only in SUBTR.

RSMIN - minimum residual reduction allowed in filtering process in IGN for a particular eigenshift.

RZ(L) - the r_a of the Lth distance, corresponding to r_{AB} in Eq. 5

s(N) - the Nth s-value s(N) = s(1) + N*DS where s(1) is the initial value of s and DS the s increment.

SCTI - Scale factor for total intensity and standard deviation. Can be used to scale date prior to analysis.

SD(IDS) - is the value of the standard deviation after the current refinement cycle for the IDSth range of DEL.

$$SD(IDS) = \sqrt{\sum_{L=1}^{NOBS(IDS)} DEL(L)^2/NOBS(IDS)}$$

SDOLD(IDS) - is the standard deviation for the IDSth region of the least squares minimization before the current refinement cycle.

$$SDOLD(IDS) = \sqrt{\sum_{L=1}^{NOBS(IDS)} DEL(L)^2/NOBS(IDS)}$$

SF(NC) - the scale factor (SK/SK3(NC)) necessary to bring the theoretical atomic intensity to the level of the NCth data set for the subtraction in EDIL. If SF(NC) = 0.0, no atomic theory is subtracted.

SFRST(NC) - the minimum s-value for the NCth data range.

SFT - the fraction of the shift vector calculated in the least-squares refinement which will actually be applied. SFT = 1.0 gives the full shift SFT = 0.5 gives half of the shift, etc.

SG(N) - The statistical uncertainty of TI(N,NC) at s(N). For XD and ND it is the counting statistics. For ED it equals a percent uncertainty in TI(N,NC).

SH(100) - contains accumulated sum of parameter shifts.

SHIFT - maximum linear parameter shift (eigenshift) allowed in filtering process in IGN.

SIS(N,NC) - the reduced intensity for the NCth data set at s(N).

SK - An overall scale factor used to bring the theory to the same scale as the experiment. SK corresponds to SK in EQ. 7.

SK3(NC) - the scale factor for the NCth data set used to bring the various data sets to the same level. Used only for multiple sets of overlapping data or to scale experimental data to yield an SK $\simeq$ 1.

SK5(NC) - the overall scale factor of the background for the NCth data set.

SKR - A scale factor for the total intensity plot in SUBTR. If SKR > 0.0, TI(N,NC) and SKR*TI(N,NC) are both plotted.

SLST(NC) - the maximum s-value for the NCth data set.

SNN(I,NC) - the midpoint for the Ith exponential segment of the NCth data set. The first segment of the NCth data set ranges from SFRST(NC) to SNN(2,NC), the last segment of the NCth data set ranges from SNN(NCV(NC) -1, NC) to SLST (NC), and the Mth segment $^{(M\neq 1)}_{(M\neq NCV(NC))}$ range from SNN(M-1,NC) to SNN(M+1,NC).

SOL(N) - are the dependent parameter changes obtained from the independent parameter changes when constraints on the structural parameters are employed.

SS(I) - the s-values for the Ith point of the initial background for the current data range used in CURVE to generate initial background parameters.

Symmetry transformation matrix and translations vector for the Mth symmetry operation on the LTth atom in the asymmetric unit yielding transformed coordinates XAA, YAA, ZAA.

$$\begin{vmatrix} FX1(M) & FX2(M) & FX3(M) \\ FY1(M) & FY2(M) & FY3(M) \\ FZ1(M) & FZ2(M) & FZ3(M) \end{vmatrix} \begin{vmatrix} XX(LT) \\ YY(LT) \\ ZZ(LT) \end{vmatrix} + \begin{vmatrix} DX(M) \\ DY(M) \\ DZ(M) \end{vmatrix} = \begin{vmatrix} XAA \\ YAA \\ ZAA \end{vmatrix}$$

SZ - "Reduced bulk-density" - ρ_o.

$$\rho_o = \rho_o' \left[\left(\sum_{uc} f_{s=0} \right)^2 / \left(\sum_{uc} f_{s=0}^2 \right) \right]$$

where ρ_o' = bulk density in units of composition/A^3.

TI(N,NC) - the intensity for the NCth data range at the Nth s-value.

TIA(N) - the total theoretical atomic intensity at the Nth s-value.

TSDOLD - is the total standard deviation over the NHRO points of DEL before the current refinement cycle.

$$TSOLD = \sqrt{\sum_{L=1}^{NHRO} DEL(L)^2 / (NHRO-NPARR)}$$

TSIS(N) - the theoretical reduced intensity at S(N).

TSISO(N) - *the theoretical reduced intensity at* S(N) corresponding to the long distances generated in LDSIS. This array is zeroed if no long distance contributions are calculated.

U(3), V(3), W(3) - are arrays used in CURVE to indicate the s value, observed background and calculated background obtained from a 3-point fit to initial background points.

V(100) - scratch array used in IGN to calculate the product of the transposed eigenvector matrix and residual vector.

WAVENO - (2k) or ($4\pi/\lambda$) for the current data set. Used only in EDIL.

WOBS(K) - the weight of the Kth observation range.

WT - the weighting function, which is used to form the combined intensity (CSIS) from the individual intensities (SIS's) when multiple ranges are used. WT is 0.5 in an overlap and 1.0 otherwise. This combination is performed in SUBSIS. Used only when NCAS $\neq$ 1.

WT(300) - is the weighting applied to insure proper coordination number calculation in LDSIS.

WTNG(100) - is a weight vector generally equal to the reciprocal of the square root of the diagonal coefficient matrix elements. It is used to normalize the coefficient matrix diagonal elements to unity.

WT1,WT2 - The weighting functions used to combine the exponential segments to obtain the background for the NCth data set. WT1 = WT2 = 0.5 for overlapping segments and WT1 = 1.0 in the non-overlap regions. WT1 and WT2 correspond to $W_m(s)$ in EQN. 8. This combination is performed in SUBBG and SUBSIS.

X(N1,N1) - eigenvector matrix is obtained from EIGRS in this array.

XCHG(I) - is the change calculated from the current refinement cycle for the Ith parameter.

XLAB - the amplitude of vibration for the current short distance. The B-value is calculated by B(L) = -XLAB × XLAB/2.0.

XNEW(I) - is the new value of the parameter I after the current cycle.

XOLD(I) - is the value of parameter I before the current refinement cycle.

XSD(I) - is the standard derivation for the Ith parameter

$$XSD(I) = \sqrt{AK(I,I)*DEUW}$$

where AK is the inverse matrix.

XX(I,J) and YY(I,J) are the scratch arrays used in the call to the plotting routine CCPLT to represent the abscissa and ordinate of the Ith point of the Jtn curve.

XX(300) YY(300) ZZ(300)- are the fractional coordinates of the atoms in the asymmetric unit in LDSIS.

YSD(N) - are the dependent parameter standard derivations obtained from the independent parameter deviations by the transformation matrix QINV.

Z(100) - is a rejection diagonal matrix. It has a value of 1 for an eigenvalue which will be accepted and zero otherwise.

IV. PLOTTING PACKAGE - PLOTT

A. Purpose

The purpose of this subroutine is to obtain directly on a line printer a crude plot. The subroutine will plot any given number of arrays on a variable size surface limited only by the printer capacity. The subroutine does not disturb the arrays being plotted. Alternate entry points allow for plotting every 2nd, 3rd or 5th point, or a calcomp plot.

B. Usage of the Program

1. Operational Procedure: Usable as a subroutine.

2. Input data: The calling sequence is:

SUBROUTINE PLOTT (XX,YY,NDATA,NDMAX,ISYMBOL,NF,
XLINE,MX,YLINE,MY,NLS,NCL,MM,LL,AREA,YSCALE).

where,

XX(J,I) and YY(J,I) are the coordinate vectors of the j-th point of the I-th array to be plotted. (e.g. I=1, to plot one array).

NDATA(J) is the number of points on the J-th array.

NDMAX is the maximum number of points in the largest array to be plotted.
In the dimension statement:
XX(J,I) and YY(J,I): J must be the value NDMAX.
(e.g. NDMAX = 100, see NDATA(J))

ISYM(J) is the symbol used in the J-th array. The ISYMBL array may be defined by a Hollerith statement (e.g. ISYM (1) = "*").

NF is the number of arrays to be plotted. In the dimension statement:
XX(J,I), YY(J,I), NDATA(I), and ISYMBL(I):
I should be equal to or greater than the value NF. (e.g. NF = 1, to plot one array)

XLINE(I) is the value for the I-th x-reference line. (e.g. leave undefined).

MX is the number of x-reference lines. In the dimension statement:
XLINE(I): I should be equal to or greater than the value MX. (e.g. MX = 0, if no x-reference lines are desired).

YLINE(I) is the value for the I-th y-reference line. (e.g. leave undefined).

MY is the number of y-reference lines. In the dimensions statement:
YLINE(I): I should be equal to or greater than the value MY. (e.g. MY = 0, if no y-reference lines are desired).

NLS is the number of lines used in the plot. In the dimension statement:
AREA(30,J) and YSCALE(J): J should be equal to or greater than the value NLS. (e.g. NLS = 56, for one page). If NLS<0, the tic marks of the calcomp plot will not be labeled. The x-dimension of a calcomp plot is NLS/10 inches.

MM control integer: MM = 1 plots across the page: MM = 2 plots down the page.

LL control integer: LL = 1 linear, LL = 2 semi-log, LL = 3 square root.

AREA(30,J) is the vector which spans the plot surface. This vector is in the call list to satisfy the variable dimensioning requirements.

YSCAL(J) is the scale scalar running down the page. This scalar is in the call list to satisfy the variable dimensioning requirements.

3. Output Format: Not necessary for user to provide output format.

4. Caution to User: The number of lines (NLS) drastically alters the storage needed in plotting because of the array AREA(30,NLS). NLS = 56 usually is near to exhausting a page of output. In cases where the user wishes

20 or more pages in one plot the dimension of AREA will be significant.

The user must alter the restrictive statement in PLOTT if NLS is needed to be greater than 512, if MX is needed to be greater than 32, or if NF is needed to be greater than 16. The restrictions are only intended to prevent unknowledgeable programmer errors.

5. Error Provision: Error stops are printed in output.

Most previously used problems have been that the values for variables in the CALL list are not what the programmer intends. Print out all variables in the CALL list as a first step in seeking errors and check list against SUBROUTINE list.

6. Alternate entry points: All entry points have the same parameter list.

PLOTT2 - will plot every 2nd point of each vector.
PLOTT3 - will plot every 3rd point of each vector.
PLOTT5 - will plot every 5th point of each vector.
CCPLT - will also yield a calcomp plot of dimensions (NLS/10) "by 10" and by making NLS and NCL negative the labels of the axis tie marks are omitted.

7. Subprograms Required: Not applicable.

8. Accuracy: The plotting accuracy is limited by the discreteness of the print spacing or the precision of the calcomp plotter.

C. Mathematical Method

The maximum and minimum values of each coordinate are found. The data are than transformed by an appropriate transformation such that the values range from 0 to "NCL" for the horizontal coordinate and from 0 to "NLS" for the vertical coordinate. These are rounded and truncated into integer values. The integer values are then used to locate the Hollerith symbol in memory. If this location is already occupied by a symbol, an equal sign is substituted for the symbol. The entire

Hollerith array than represents the plots and is consequently printed.

For calcomp plotting, it is assumed that the arrays to be plotted will be ordered into either increasing or decreasing values of the abscissa.

D. Example: For a $15A^{-1}$ plot at $1A^{-1}$/INCH, NLS = 150 XLINE(1) = 0, XLINE(2) = 15.0

The program will put out both a line printer and calcomp plot.

VII. LEAST-SQUARES METHOD

SYMBOLS:

V - function to be minimized.

F - a function of the x dependent variables describing the observational parameters.

F^O - observed value of F.

F^C - calculated value of F.

$\Delta F-(F^O-F^C)$ - the residual.

D - the derivative matrix $D_{ij} = \partial F_i/\partial x_i$ for the ith observation and the jth parameter.

W_i - weight for the ith observation.

A - the coefficient matrix D^tD.

Δx - the shift vector.

$\Delta x'$ - normalized parameter change.

$\Delta x''$ - normalized eigenshifts (linear combination parameter changes).

B - residual vector weighted by derivatives D^tF.

N - normalization weight matrix $1/\sqrt{A_{ii}}$

T - eigenvector matrix.

t - transpose operation.

λ - eigenvalue.

x - dependent parameters.

v - independent parameters.

z - filtering diagonal matrix of ones and zeros.

- total standard deviation.

s - standard deviation for parameters.

S - correlation matrix.

E - reduced independent derivative matrix

$$E_{ik} = \frac{\partial F_i}{\partial v_k} = \sum_j \frac{\partial F_i}{\partial x_j} \cdot \frac{\partial x_j}{\partial v_k}$$

m - number of constraint

M - number of observations.

n - number of unconstrained dependent variables.

k - number of independent variables.

Matrix Equations

$$V = \sum_{i=1}^{M} W_i (F_i^O - F_i^C)^2$$

We wish to minimize this equation

$$\frac{\partial V}{\partial x_j} = 0 = \sum_{i=1}^{M} W_i \ (F_i^O - F_i^C) \ \frac{\partial F_i^C}{\partial x_j} \qquad (j=1,2\ldots n)$$

This gives the set of n normal equations in n unknowns.

When F^C is not linear it must be approximated by a Taylor series in Δx where higher order terms are neglected

$$F(x_1,x_2\ldots x_n) = F(a_1,a_2\ldots a_n) + \frac{\partial F(a_1,a_2\ldots a_n}{\partial x_1} \ \Delta x_1 + \ldots$$

$$\ldots + \frac{\partial F(a_1,a_2,\ldots a_n)}{\partial x_n} \ \Delta x_n$$

When the a_j's are good approximations, application of the least squares will yield values for the parameter changes Δx_j, where

$$x_{jnew} = x_{jold} + \Delta x_j$$

In matrix notation the normal equations can be written as

$$A\Delta x = B$$

where

$$a_{kj} = \sum_{i=1}^{m} W_i D_{ij} \ D_{ik}$$

$$D \equiv d_{ij} = \partial F_i^C / \partial x_j$$

$$B \equiv b_k = \sum_{i=1}^{m} W_i \Delta F_i \ D_{ij}$$

$$A = D^t D \qquad B = \Delta F D^t$$

When m constraints are applied on the ,n, dependent parameters, x, we reduce the number of variables to k (n-m) independent parameters v. The normal equations are now

$$A\Delta v = B$$

A and B must be recalculated in terms of the original derivative matrix D. The new derivative matrix E, in terms of the K independent variable v is given by

$$E = J^t D$$

where J^t is a transformation matrix relating the derivatives of the dependent parameters with respect to the independent parameters. The derivative of F^c with respect to the independent parameters is obtained by the chain rule.

$$A = E^t E$$

$$B = \Delta F \; E^t$$

The dependent solutions are given by

$$\Delta x = J \Delta v$$

The parameter change vector can be obtained by matrix inversion or the eigenvector-eigenvalue method

1. Matrix Inversion (INVERT)

$$A \; \Delta x = B$$

$$(NAN) \; (N^{-1} \; \Delta x) = N \cdot B$$

$$\Delta x' = N^{-1} \; \Delta x = (N A N)^{-1} \cdot (NB)$$

$$\Delta x = N \cdot \Delta x'$$

2. Eigenvector-eigenvalue (IGN):

$$A\ \Delta x = B$$

$$(NAN)\ (N^{-1}\ \Delta x) = NB$$

$$T^{-1}\ (NAN)\ T\ T^{-1}\ (N^{-1}\Delta x) = T^{-1}\ (NB)$$

$$\lambda = T^{-1}\ (NAN)\ T$$

$$T^{t} = T^{-1}$$

$$\lambda \cdot T^{-1}\ (N^{-1}\ \Delta x) = T^{t}\ (NB)$$

$$\Delta x'' = T^{-1}\ (N^{-1}\ \Delta x) = z \cdot \lambda^{-1} \cdot T^{t}\ (NB)$$

$$\Delta x = N\ T\ x''$$

$$\sigma = \left[\sum_{i=1}^{M} \Delta F^{2}/M\text{-}n + m\right]^{1/2}$$

Standard deviation of the J^{th} parameter

$$S_j = \sigma\sqrt{A_{jj}^{-1}}$$

Correlation matrix

$$S_{ij} = A_{ij}^{-1} / \sqrt{A_{jj}^{-1}\ A_{ii}^{-1}}$$

VIII. Program package consists of:

1. This users manual.
2. A magnetic tape with Fortran coding and input/output files for an electron diffraction analysis of silicon and an x-ray diffraction analysis of silica glass.
2. A printed listing of these files.

marks of the calcomp plot will not be labeled.
The x-dimension of a calcomp plot is NLS/10
inches.
ontrol integer: MM = 1 plots across the page:
MM = 2 plots down the page.
ontrol integer: LL = 1 linear, LL = 2 semi-log,
LL = 3 square root.
(30,J) is the vector which spans the plot
surface. This vector is in the call list to
satisfy the variable dimensioning requirements.
L(J) is the scale scalar running down the page.
This scalar is in the call list to satisfy
the variable dimensioning requirements.
ecessary for user to provide output format.
number of lines (NLS) drastically alters
storage needed in plotting because of the array
(30,NLS). NLS = 56 usually is near to exhausting
ge of output. In cases where the user wishes